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AUTHOR(S): THOMAS R. BEMENT
AND
JOHN R. PHILLIPS

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University of California

LOS ALAMOS SCIENTIFIC LABORATORY
Post Office Box 1663 Los Alamos, New Mexico 87545
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Evaluation of Relative Detection Efficiency on Sets of Irradiated Fuel Elements

by

Thomas R. Bement
and
John R. Phillips

Los Alamos Scientific Laboratory
Los Alamos, New Mexico 87545

ABSTRACT

The accurate analysis and monitoring of the nuclear material inventory of discharged fuel elements is a critical part of the total system of safeguards for the nuclear fuel cycle. One of the nondestructive techniques used for verifying irradiated fuel elements is high-resolution gamma spectrometry. One aspect of this technique is the evaluation of the relative detection efficiency of gamma-ray detection systems. Many conditions, including the modification of fuel elements, can affect the relative detection efficiency. Assuming no detector malfunctions, relative detection efficiency at a fixed energy level should be a constant (within measurement uncertainty) for all fuel elements having the same irradiation history. An application of a Hotelling's - T^2 type statistic is presented as a method of screening measurements performed on large sets of irradiated elements and identifying possible outliers.

I. INTRODUCTION

The Nuclear Nonproliferation Treaty (NPT) requires the signatories to establish a national material accountancy system for special nuclear materials (SNM).[1] This system and SNM inventory must be verified periodically by the International Atomic Energy Agency (IAEA) as part of the required safeguards system. Most significant quantities of SNM are found in either fuel elements in operating reactors or in spent-fuel elements in storage facilities. As part of the safeguards inspection, the operator-declared values for the fissile material (^{235}U, ^{239}Pu, ^{241}Pu) remaining in spent-fuel elements must be verified. The most widely accepted technique is the use of high-resolution gamma spectrometry (HRGS) to measure the relative concentrations and ratios of fission products.[2] These isotopic ratios are correlated with the declared exposures of the fuel elements.[3] If the exposure can be verified by the inspectors, then the remaining fissile content can be calculated using isotopic correlation techniques.[2]

The usefulness of the HRGS measurement technique can be limited by the source self-attenuation. For example, in a Pressurized Water Reactor (PWR) fuel assembly, less than 6 percent of the 1500 keV gamma rays originating in the center rods can reach the surface. However, this factor (source self-attenuation) that limits the usefulness of HRGS can be used to obtain additional information about the fuel element. If the fuel element has been modified by removal of irradiated material or by substitution of material, then the observed relative intensities of gamma rays originating from the same isotope will be changed.

Define the relative detection efficiency for a specific measurement geometry and at a particular energy level as the relative probability of a gamma ray escaping the fuel material, passing through any absorbing material and producing a pulse in the full-energy peak. The relative detection efficiency function is then determined by dividing each full-energy peak area by

its gamma-branching yield and expressing this ratio as a function of energy level.[4-6] (Gamma branching yields are physical constants and are the relative intensities of gamma rays originating from the same isotope.) This relative efficiency function is essentially the product of the detector efficiency and the sample attenuation expressed as a function of gamma-ray energy. Thus, changes in the efficiency function could be indicative of changes in self-attenuation, which in turn might point to fuel element modification by source removal or substitution. These diversion scenarios could possibly go undetected with the usual application of HRGS because the operator could simply declare slightly lower exposures to explain the lower measurements caused by a diversion.

We will discuss a method for screening HRGS data based on changes in the measured relative detection efficiencies that can be easily applied to data as it is collected at the spent-fuel storage facility. The method flags fuel elements that do not have a relative detection efficiency function consistent with other members of the set. If a physical reason for the observed discrepancy is not evident (e.g., a change in scanning geometry such as the introduction of an absorber in the gamma-ray beam), these elements can then be examined in greater detail to ensure the verification of the operator-declared exposure values.

II. EXPERIMENTAL MEASUREMENTS

One hundred thirty-seven fuel elements from a heavy-water research reactor were measured using HRGS. These fuel elements contained natural uranium metal as fuel and were irradiated to exposures ranging from 100 to 1000 megawatt days per metric ton uranium (MWd/tU). A single gamma-ray spectrum (345-2433 keV) was collected at the central axial position. Data from the isotopes ^{134}Cs, ^{144}Pr, and ^{106}Rh were used in the screening procedure.

Table I lists the gamma-ray energies and relative branching yields for each of the three isotopes.

TABLE I

GAMMA-RAY ENERGIES AND BRANCHING YIELDS

Isotope	Energy (keV)	Relative Branching Yield[7,8,9]
^{134}Cs	604.6	0.976
	795.8 + 801.8	0.941
	1365.1	0.0304
^{144}Pr	696.5	0.0135
	1489.1	0.0027
	2185.7	0.0067
^{106}Rh	621.8	0.0975
	873.1	0.00414
	1050.1	0.015
	1128.0	0.00383
	1562.0	0.0015

Table II lists the relative intensities for the energies in Table I from a typical fuel element with a medium exposure value and from one with a low exposure value. Percent relative standard deviations (the assumed standard deviation of the distribution from which the observation was taken, expressed as a percentage of the observation) are also given.

TABLE II

EXAMPLE OF HRGS DATA AND RELATIVE ERRORS

DECLARED EXPOSURE (MWd/tU)	ISOTOPE	GAMMA-RAY ENERGY (keV)	RELATIVE INTENSITY	PERCENT RELATIVE STANDARD DEVIATION
545.76	^{134}Cs	604.6	0.1867	2.45
		795.8 + 801.8	0.4448	1.21
		1365.1	0.0260	7.32
	^{144}Pr	696.5	0.5947	0.94
		1489.1	0.3773	0.94
		2185.7	0.9341	0.64
	^{106}Rh	621.8	0.5686	1.06
		873.1	0.0705	4.03
		1050.1	0.2902	1.21
		1128.0	0.0809	3.24
		1562.0	0.0430	4.23
152.78	^{134}Cs	604.6	0.0186	16.25
		795.8 + 801.8	0.0272	9.84
		1365.1	0.0019	54.25
	^{144}Pr	696.5	0.0249	1.55
		1489.1	0.1374	1.36
		2185.7	0.3327	0.81
	^{106}Rh	621.8	0.1270	2.63
		873.1	0.0118	13.41
		1050.1	0.0651	2.75
		1128.0	0.0181	7.98
		1562.0	0.0109	9.41

III. DEVELOPMENT OF THE SCREENING PROCEDURE

In this section, we propose a procedure for screening a large set of fuel elements for ones that have relative detection efficiency functions that are not consistent with other members of the set. The form of the efficiency function is unknown, so the procedure will be based on ratios of gamma-ray intensities. The model for observed gamma-ray intensity is based on the assumption that the expected value of the intensity is the product of the actual intensity, the detector efficiency, and the appropriate branching yield. Define

Y_{ijm} = relative intensity of the m^{th} gamma ray, j^{th} isotope, i^{th} fuel element, $i=1,\ldots,n$; $j=1,\ldots,t$; $m=1,\ldots,p_j$. Assume Y_{ijm} and $Y_{i'jm}$ are independent for $i' \neq i$.

x_{jm} = energy level of the m^{th} gamma ray, j^{th} isotope.

$f(x_{jm})$ = relative detection efficiency at energy level x_{jm}.

B_{jm} = branching yield for the m^{th} gamma ray, j^{th} isotope (physical constant).

D_{ij} = relative disintegration intensity of the j^{th} isotope, i^{th} fuel element.

θ_{ijm} = coefficient of variation of Y_{ijm} (assumed known).

The expected value of Y_{ijm} can now be written in terms of the efficiency function, f, as

$$E(Y_{ijm}) = B_{jm} D_{ij} f(x_{jm}) \quad ,$$

and the variance of Y_{ijm} is

$$Var(Y_{ijm}) = \theta^2_{ijm} E^2(Y_{ijm}) \quad .$$

Note that in this model, the function f is assumed to be the same for all fuel elements under consideration. As mentioned previously, this assumption will not be satisfied in cases where fuel elements have been modified, and this is the reason for searching for anomalous efficiency functions.

A method of identifying fuel elements with potentially inconsistent efficiency functions will now be developed. Consider the ratio of intensities of two gamma rays from the same isotope. Using error propagation, one can obtain

$$E\left(Y_{ijm}/Y_{ijm'}\right) \approx \left(1 + \theta^2_{ijm'}\right) \frac{B_{jm} f(x_{jm})}{B_{jm'} f(x_{jm'})} \qquad (1)$$

and

$$Var\left(Y_{ijm}/Y_{ijm'}\right) \approx \left(\theta^2_{ijm} + \theta^2_{ijm'}\right)\left(\frac{B_{jm} f(x_{jm})}{B_{jm'} f(x_{jm'})}\right)^2 \quad . \qquad (2)$$

An important thing to note about Eq. (1) and (2) is that, except for the θ-terms, the right-hand sides are constant over all fuel elements. Because of this, the values of the branching yields and the true form of the efficiency function will be seen to be unimportant in the development of the screening procedure.

Only selected ratios are used for each isotope. The choice of gamma-ray ratios and, in particular, the choice of the denominators was made to enhance the accuracy of the propagations.

Let Q_{ijk} denote the k^{th} ratio for the j^{th} isotope and i^{th} fuel element ($i=1,\ldots,n$; $j=1,\ldots,t$; $k=1,\ldots,r_j$). Also, we can rewrite Eq. (1) and Eq. (2) as

$$E(Q_{ijk}) = a_{ijk}\mu_{jk}$$

and

$$Var(Q_{ijk}) = d_{ijk}\mu_{jk}^2 \quad ,$$

where $a_{ijk} = 1 + \theta_{ijm'}^2$, $d_{ijk} = \theta_{ijm}^2 + \theta_{ijm'}^2$, and m and m′ are the indices of the gamma rays involved in the ratio Q_{ijk}. Similarly,

$$\mu_{jk} = \left[B_{jm} f(x_{jm})\right] / \left[B_{jm'} f(x_{jm'})\right] \quad .$$

Except for a_{ijk} and d_{ijk} (which are assumed to be known because the θ-terms are assumed to be known), this mean and variance are the same for all fuel elements. Also, covariance terms are of the form

$$Cov(Q_{ijk}, Q_{ijk'}) = \rho_{kk'}\mu_{jk}\mu_{jk'}\sqrt{d_{ijk}d_{ijk'}} \quad ,$$

where $\rho_{kk'}$ is unknown but is constant over all fuel elements. The only part of this expression that is not constant over all fuel elements is the factor under the radical and it is known.

The next step will be to transform the ratios so that their means, variances, and covariances are the same for all fuel elements. Define

$$U_{ijk} = Q_{ijk}/a_{ijk} \quad .$$

Then

$$E(U_{ijk}) \approx \mu_{jk}$$

and

$$Var(U_{ijk}) \approx \frac{d_{ijk}}{a_{ijk}^2}\mu_{jk}^2 \quad .$$

That is, within the limits of propagation accuracy, the U_{ijk} have a constant mean over all fuel elements. Let

$$\bar{U}_{jk} = \frac{1}{\sum w_{ijk}} \sum_{i=1}^{n} w_{ijk} U_{ijk} \quad ,$$

where $w_{ijk} = a_{ijk}^2 / d_{ijk}$. Let

$$V_{ijk} = \left(U_{ijk} - \bar{U}_{jk}\right)\left(1/w_{ijk} - 1/\sum_{i=1}^{n} w_{ijk}\right)^{-1/2}$$

$$\approx \left(U_{ijk} - \bar{U}_{jk}\right) \sqrt{w_{ijk}}$$

for large sample sizes.

Then

$$E(V_{ijk}) \approx 0 \quad ,$$

$$\mathrm{Var}(V_{ijk}) \approx \nu_{jk} \quad ,$$

and

$$\mathrm{Cov}(V_{ijk}, V_{ijk'}) \approx \rho_{kk'} \nu_{jk} \nu_{jk'} \quad .$$

The V_{ijk} and $V_{ijk'}$ are correlated because they all have $\bar{U}_{jk}$ in common. This correlation decreases however as Σw_{ijk} increases, and it will be ignored here since we have a large number of fuel elements.

Let $\underset{\sim}{V}_i'$ be the row vector of V_{ijk} terms for the i^{th} fuel element,

$$\underset{\sim}{V}_i' = (V_{i11}, \ldots, V_{i1r_1}, \ldots, V_{it1}, \ldots, V_{itr_t}) \quad .$$

Then

$$E(\underset{\sim}{V}_i) = \underset{\sim}{0}$$

and

$$\mathrm{Var}(\underset{\sim}{V}_i) \approx \underset{\sim}{\Sigma}, \; i=1, \ldots, n \quad ,$$

where $\underset{\sim}{\Sigma}$ is the same for all fuel elements. Now, if $\hat{\underset{\sim}{\Sigma}}$ is the usual sample covariance matrix and if the V_{ijk} are approximately normally distributed, then $T_i^2 = \underset{\sim}{V}_i' \; \hat{\underset{\sim}{\Sigma}}^{-1} \; \underset{\sim}{V}_i$ is distributed approximately as Hotelling's T^2, and

$$F_i = \frac{n-r}{r(n-1)} T_i^2$$

is distributed approximately as an F with r and n-r degrees of freedom, where $r = \sum_{k=1}^{t} r_k$ and n is the number of fuel elements.

The screening process now consists simply of computing F_i for each fuel element and flagging those elements where F_i exceeds some critical value.

The significance level associated with this screening procedure would be difficult to determine because of the approximations that have been made and the effect sample size has on them. However, Beckman and Whiteman[10] have developed a test statistic based on generalized distances for testing for an arbitrary number of multivariate outliers, and it can be used to give a crude significance level in this case. They give critical values for up to five outliers with sample sizes of 100 or less. Their method as applied to the screening process works as follows. Let $F_1,\ldots,F_n$ be the computed F values ordered from smallest to largest. To test for at most k outliers, compare F_{n-k+1} with a critical value, CV(k,1). If F_{n-k+1} is larger, the elements corresponding to $F_{n-k+1},\ldots,F_n$ are declared outliers. If F_{n-k+1} is smaller, then F_{n-k+2} is compared with another critical value, CV(k,2), and so on.

Many assumptions and approximations have been used in the development of this screening procedure. Some of these points will now be addressed.

The mean and variance given in Eq. (1) and (2) are approximations determined by error propagation. Simulation studies indicate that for typical

data, the relative error of propagation for both the mean and variance can be kept less than 5% by proper selection of ratios. In particular, one should avoid using gamma rays with low intensities (and high relative standard deviations), especially in the denominator. The simulations were based on samples of 500 and the assumption that the intensities are normally distributed with relative standard deviations less than 0.15.

As mentioned earlier, V_{ijk} and $V_{ij'k}$ are correlated. However, it can be shown that the covariance approaches zero as $\sum_{i=1}^{n} w_{ijk}$ increases. Now, $w_{ijk} = (1+\theta_{ijm}^2)^2/(\theta_{ijm}^2+\theta_{ijm'}^2)$. (m and m′ are the indices of the gamma rays involved in the k^{th} ratio.) If energy levels with relative standard deviations of 0.15 or less are used (i.e., all $\theta_{ijk} \leq 0.15$), then $w_{ijk} \geq 23$ and hence the covariance is $O(1/n)$. Thus, the assumption that V_{ijk} and $V_{ij'k}$ are uncorrelated is reasonable for large sample sizes. We have not yet investigated the case of a small sample size.

The assumption that the V_{ijk} are normally distributed can certainly be questioned. The ratio of two normals (assume the denominator distribution is truncated below some positive value) is not normal. However, as the variance of the denominator decreases, the distribution of V_{ijk} approaches that of a normal. With this in mind, data can be collected and ratios constructed so that the denominator variances are kept as small as possible. Mardia[11] states that the true significance level of the Hotelling's T^2 one-sample test is larger than one would expect when data are not multivariate normal. The extent of the nonnormality of the ratios and its effect on the significance level of the screening procedure has not yet been investigated.

IV. RESULTS

The screening procedure was applied to the gamma-ray spectra of the 137 fuel elements discussed in Sec. II. The ratios that were used are given in

Table III. Both ordinary and log-transformed ratios were used and no appreciable differences were observed between the two sets of results. The following discussion is based on the analysis of the nontransformed ratios.

TABLE III

GAMMA-RAY RATIOS USED FOR SCREENING PROCEDURE

^{134}Cs GAMMA-RAY RATIOS	^{106}Rh GAMMA-RAY RATIOS	^{144}Pr GAMMA-RAY RATIOS
796/605	873/622	1489/696
1365/605	1050/622	2186/696
1365/796	1128/622	1489/2186
	1562/622	
	873/1050	

Table IV lists the F values that were computed for each of the 137 fuel elements. The 5% and 1% critical values (CVs) for an F with 14 and 123 degrees of freedom are 1.77 and 2.23, respectively. Elements with F values that exceed these CVs are indicated. Elements 39, 40, 41, 61, 90, and 106 exceed the 1% CV. In addition, elements 42, 49, 55, 65, 67, 80, 111, and 116 have F values that exceed the 5% CV. Examination of field records showed that the computer code used to determine peak areas (gamma intensities) was modified for elements 39, 40, and 61 because of the extremely low activity for the 605 keV gamma ray of ^{134}Cs. These were the only three spectra treated differently and they were all identified by the screening process. Elements 41, 49, 67, and 90 had very low exposure values (150 MWd/tU) and consequently the relative errors associated with ratios for these elements were large. No explanation based on the experimental measurement conditions could be found for the remaining fuel elements that were flagged by the screening process.

TABLE IV

F VALUES FOR 137 ELEMENTS

Element	F	Element	F	Element	F	Element	F
1	1.14242	35	0.14555	69	0.55680	103	0.68276
2	0.77092	36	1.45604	70	0.35332	104	1.76548
3	0.83796	37	0.61129	71	0.74494	105	0.43836
4	1.08480	38	1.13926	72	0.66157	106	2.39220**
5	0.58495	39	2.52359**	73	0.41660	107	1.71084
6	1.61501	40	5.15594**	74	1.19028	108	0.60129
7	0.92377	41	2.92583**	75	1.19118	109	0.34207
8	0.42744	42	1.83320*	76	0.60979	110	1.48761
9	0.47271	43	0.46360	77	0.72536	111	2.10164*
10	0.16563	44	1.02429	78	0.53141	112	0.58676
11	0.64410	45	1.65681	79	0.06514	113	1.33118
12	0.38352	46	0.25449	80	1.80001*	114	0.94786
13	0.58211	47	0.67774	81	0.10867	115	0.73951
14	0.64700	48	1.46201	82	0.77727	116	2.22658**
15	0.47341	49	1.89577*	83	0.46932	117	0.80387
16	0.65966	50	1.52005	84	0.94964	118	0.37526
17	0.32876	51	1.38614	85	0.79735	119	0.51960
18	0.38188	52	0.45366	86	0.56760	120	0.77757
19	0.20992	53	0.48270	87	0.58993	121	1.05880
20	1.57703	54	0.80580	88	1.25143	122	0.91992
21	0.32775	55	2.19833*	89	0.18618	123	0.38637
22	0.49465	56	0.44028	90	2.51296**	124	1.09311
23	0.77093	57	0.40444	91	0.33358	125	0.70299
24	0.52175	58	0.49172	92	0.87805	126	0.97719
25	0.93327	59	0.29230	93	0.47654	127	0.72248
26	0.39247	60	0.81450	94	1.01023	128	0.25368
27	0.51771	61	3.17440**	95	0.42324	129	0.63037
28	1.66054	62	0.77633	96	0.91423	130	0.47226
29	0.94284	63	0.64192	97	0.33309	131	0.40028
30	0.35253	64	1.10313	98	0.35983	132	0.34065
31	0.86523	65	1.97854*	99	0.73672	133	1.09107
32	0.57358	66	0.34183	100	0.64772	134	0.45578
33	1.39013	67	2.11014*	101	0.78876	135	0.65222
34	1.15522	68	0.18522	102	0.96930	136	0.74760
						137	0.26360

* Exceeds $F_{0.95}$
** Exceeds $F_{0.99}$

V. CONCLUSIONS

It appears that the screening process presented here may be a practical tool that can be used by inspectors in verifying the status of irradiated fuel elements. Many types of measurements can be performed on a set of irradiated fuel elements with this technique being used to identify elements that should be examined more closely. This could improve the efficiency of an inspector by reducing the number of detailed examinations. The screening technique can be applied to sets of measurements in which the geometry is held constant. If the geometry is changed, additional fuel elements may be flagged. In this case, the procedure was successful in identifying the only fuel elements in the set of 137 that were known to have been treated differently (i.e., 39, 40, and 61), and the number of "false alarms" was not excessive.

The Beckman-Whiteman[10] critical value for five outliers was extrapolated and CV(5,1) was estimated to be approximately 1.70. Taking this as the critical value, there are 14 elements with larger F values. Seven of these may be genuine outliers as explained above, leaving seven that exceed CV(5,1). The fact that this is a larger number than one would expect may be explained in part by the previously mentioned results of Mardia[11]. In any case, more work is required if the significance level of the screening process is to be determined accurately.

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